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Optical Property and the Relationship between Resistivity and Surface Roughness of Indium Tin Oxide Thin Films

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Abstract

Indium tin oxide (ITO) films with high visible transmittance were fabricated on polymethyl methacrylate (PMMA) flexible substrate at room temperature by radio frequency (RF) magnetron sputtering. The deposition rate, microstructure, surface roughness, electrical and optical properties of these films have been investigated as a function of Ar partial pressure from 0.2-1.4Pa. The microstructure and surface morphology of ITO films were studied using X-ray diffraction (XRD) and atomic force microscopy (AFM), the resistivity was investigated by four-point probe technology, and the optical transmittance was determined by UV-Vis spectrophotometer. The results show that the resistivity of the ITO film decreases with increasing Ar partial pressure from 0.2Pa to 0.8Pa, and then increases with increasing Ar partial pressure from 0.8Pa to 1.4Pa. The resistivity varied from 1.25×10^{-3} Ω.cm to 2.33×10^{-3} Ω.cm and the average transmission in the visible range was 90%. It was also found that the microstructure is amorphous and the surface roughness decreases from 1.438nm to 0.813nm with increasing Ar partial pressure to 1.1Pa. The most interesting fact is that the resistivity increases with the increasing surface roughness, which indicates that the surface roughness also plays an important role in electrical properties of the ITO films.

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Keywords: ITO films; Transmittance; Surface roughness; Resistivity

1. Introduction

ITO thin film is a highly degenerate n-type semiconductor, which has a low electrical resistivity of $2\sim4\times10^{-4}$ Ω.cm. Furthermore, ITO is a wide band gap semiconductor (Eg : 3.5-4.3 eV), which shows high transmission in the visible and near-IR regions of the electromagnetic spectrum. Due to these unique properties, ITO has been used in a wide range of applications. For example, currently ITO films have been widely utilized as an essential part of many

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optoelectronic devices [1-4]. There is also considerable interest in plastic substrates in the optoelectronic industry, flexible display and flexible electronics [5-7]. The replacement of glass substrates with flexible substrates offers many advantages such as lightweight, toughness, and flexibility. Style or design freedom of flexible substrates allows roll-up displays and roll-to-roll manufacturing.

There are several deposition techniques used to grow ITO thin films including chemical vapor deposition (CVD), evaporation and spray pyrolysis. Recently, reactive magnetron sputtering in an Ar atmosphere offers a number of advantages for ITO manufacturing compared to deposition techniques as evaporation, chemical vapor deposition (CVD) or spray pyrolysis. High quality films can be deposited on large substrates at low temperature and at high deposition rates [8-9].

Izumi [10] studied the relation between electrical and structural properties of ITO films prepared by pulsed laser deposition with and without in situ laser irradiation, the results indicated that the ITO films prepared with in situ laser irradiation showed very low resistivity ($<10^{-4}$ Ω.cm) which can be attributed to the high crystallinity and low residual stress. Kim [11] studied the properties of ITO films as a function of oxygen partial pressure. However, the effect of surface morphology or surface roughness on the resistivity has not been reported in previous work. So, the attractive work in this paper will focus on the relationship between the surface roughness and the resistivity.

The main aim in this article is to report a study of the optical, electrical, and structural properties of ITO films deposited by RF magnetron sputtering on PMMA substrate at low temperature without a postdeposition anneal. Film properties were investigated as a function of Ar partial pressure, and the relationship between resistivity and surface roughness was also been discussed.

2. Experimental details

ITO thin films with thickness 150nm were deposited on PMMA substrates by RF sputtering at room temperature. The sputtering target used in this experiment was In_2O_3 containing 10 wt % SnO_2. The PMMA substrate was cleaned for 5 minutes in ethanol and dried in flowing N2 gas. The substrate-to-target distance in the chamber was 100mm. During the films deposition, the substrate was not heated in order to keep it close to the room temperature. The RF sputtering power was 80W, which maintained a different practical deposition rate at different Ar partial pressure was shown in Figure 1. The deposition rate decreases from 2.69nm/min to 1.49nm/min with increasing Ar partial pressure from 0.2Pa to 1.4Pa. As a result, the deposition time was adjusted accurately to keep the thickness of ITO films to be 150nm approximately. As a sputtering gas, the 99.99% Ar varied in the range of 0.2Pa to 1.4Pa was controlled by a mass flow controller. The target was pre-sputtered by Ar ions to remove its surface contamination, which may have been formed during system venting.

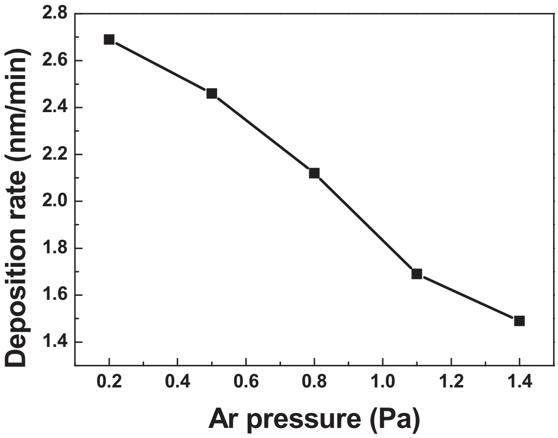


Fig.1. The deposition rate at different Ar partial pressure

The thickness of the films was determined using XP-2 stylus profiler, the crystallographic structure was determined from x-ray diffraction (XRD) with Cu-Kα anode radiation source. The surface morphology and surface roughness were examined by atomic force microscopy (AFM) with a SPA-300HV scanning probe microscopy

(SPM) system. The resistivity was measured by four-point probe technology at room temperature, five tests were performed on each sample and the average values of resistivity were calculated, and the optical transmittance in the visible region was measured using a UV-Vis spectrophotometer.

3. Results and discussion

Figure 2 shows the X-ray diffraction patterns of ITO films prepared on PMMA substrate at Ar partial pressure 0.8pa. The X-ray diffraction patterns of as-deposited ITO films did not show any peak, suggesting an amorphous phase. In addition, the as-deposited ITO films grown at other Ar working pressures also had approximately similar XRD spectra like that shown in Figure 2, and no structural discrepancies were observed between all specimens. It can be concluded that the Ar partial pressure, ranging from 0.2Pa to 1.4Pa in our experiments, had little effect on the crystal structure of ITO films due to the fact that the substrate temperature (room temperature) was below the crystallization temperature of ITO, therefore, the film grew in the amorphous phase. This result is consistent with literature reports that the phase of magnetron sputtering ITO films at low deposition temperature is amorphous [12,13].

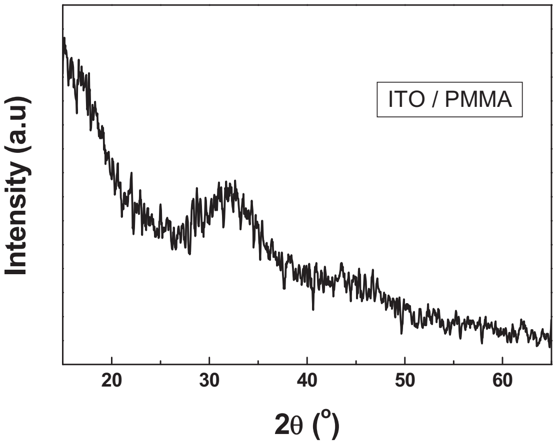
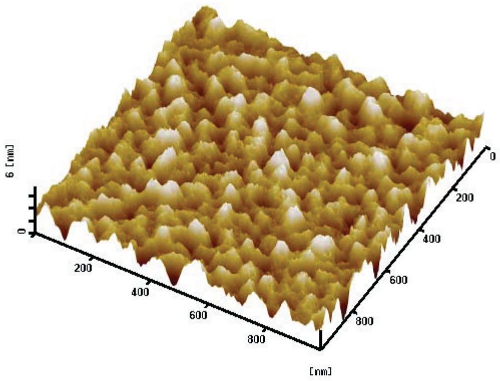


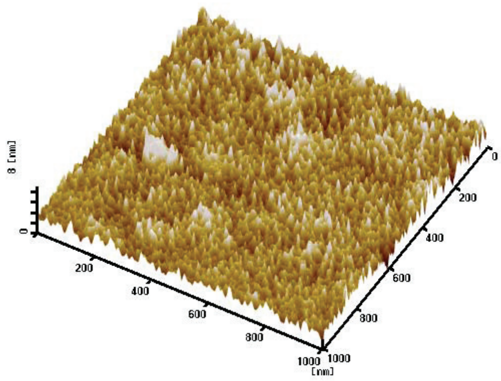
Fig.2. The X-ray diffraction patterns of ITO films prepared on PMMA substrate at Ar partial pressure 0.8pa.

Figure 3 shows the three dimensional AFM images (1×1 μm^2) of ITO film grown on PMMA substrate at different Ar partial pressure of 0.8Pa (a), 1.1Pa (b), and 1.4Pa (c), respectively. The surface roughness decreased from 0.923nm to 0.813nm with the increasing Ar partial pressure from 0.8Pa to 1.1Pa. On the other hand, the surface of the film grown on PMMA substrate at Ar partial pressure 1.4Pa exhibited a great deal of roughness 1.252nm. Three dimensional grains 80-100 nm in diameter were observed, as shown in Figure 3 (a), which would indicate typical columnar growth. The grain diameter became smaller, reducing to 50-60 nm as shown in Figure 3 (c), and it decreased further to 10-20nm at Ar partial pressure 1.1Pa as shown in Figure 3 (b). The kinetic energy of the deposited atoms during film growth should decrease with increasing Ar partial pressure, which suppressed atomic migration and reduces the growth rate of ITO. Therefore, Ar gas partial pressure during film growth is a key factor in controlling the film growth rate, leading to the improvement of surface smoothness observed in Figure 3 (b).

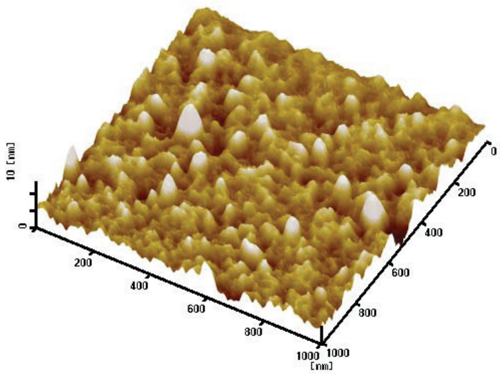
Figure 4 shows the RMS surface roughness of ITO films as a function of Ar partial pressure. The surface roughness decreased from 1.438nm to 0.813nm with the increasing Ar partial pressure from 0.5Pa to 1.1Pa. On the other hand, in our experiments, the film grown at Ar gas partial pressure 1.4Pa was non-homogeneous and some defects were observed from Figure 3 (c). The observed bigger grain structure was likely formed as a result of the gathering of very small 3D grains nucleated at many points on the substrate, which grow larger, without migration, until connected with neighboring grains. In other words, with the increasing Ar gas partial pressure from 1.1Pa, the film surface roughness increased again. The surface roughness are generally controlled by the process parameters such as working pressure, oxygen partial pressure, sputter power and substrate temperature [14-16]. In these experiments, the changes of surface roughness were due to the working Ar gas pressure since the other process parameters were consistent for the ITO film prepared.



(a) Ar partial pressure 0.8Pa



(b) Ar partial pressure 1.1Pa



(c) Ar partial pressure 1.4Pa

Fig.3. The three dimensional AFM images (1×1 μm^2) of ITO film grown on PMMA substrate at different Ar partial pressure. (a) Ar partial pressure 0.8Pa, surface roughness 0.923nm, (b) Ar partial pressure 1.1Pa, surface roughness 0.813nm, (c) Ar partial pressure 1.4Pa, surface roughness 1.252nm.

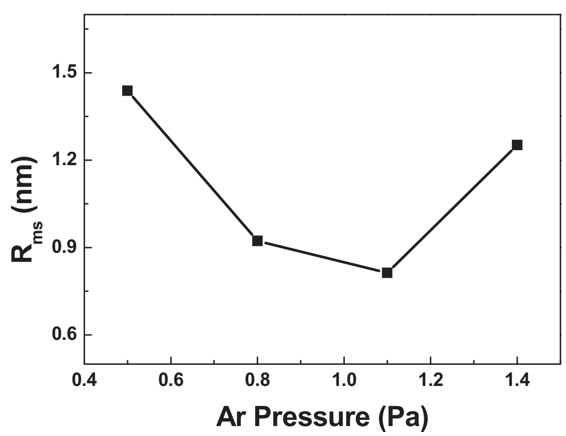


Fig.4. The RMS surface roughness of ITO films as a function of Ar partial pressure.

Figure 5 shows the resistivity of the ITO films as functions of the Ar gas partial pressure during deposition. For films grown on PMMA substrate, it can be found that the resistivity was very sensitive to the Ar partial pressure. When the lowest pressure (Ar 0.2Pa) is used, the resistivity was about 2.33×10^{-3} Ω.cm. And, with increasing pressure, the resistivity decreased to a minimum of 1.25×10^{-3} Ω.cm at Ar pressure 0.8Pa, then rose obviously to 1.515×10^{-3} Ω.cm at Ar pressure 1.4 Pa. The resistivity of these films was comparably higher than that of practical ITO films due to the amorphous structure of the films [17]. Accordingly, the ITO films in these studies exhibited general electronic properties deposited on unheated substrates by RF magnetron sputtering.

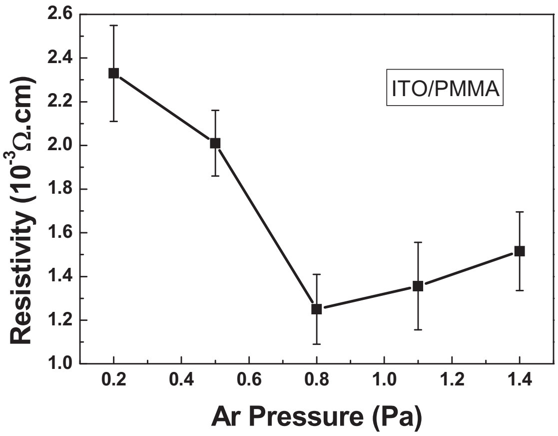


Fig.5. The resistivity of the ITO films as functions of the Ar gas partial pressure during deposition.

A similar trend in the plotted curve was found by the comparison of Figure 4 and Figure 5, which indicated surface roughness and resistivity of the ITO film were related to a certain extent. Figure 6 shows the relation between surface roughness and resistivity. The resistivity of ITO film increased with increasing the surface roughness and the variation of resistivity was a direct consequence of surface roughness, which caused electron scattering possibly due to the surface roughness of films dominated the scattering of the electrons reducing their mean free path lengths and thus increasing the resistivity. This result indicates that the surface roughness also plays an important role in electrical properties of the ITO films.

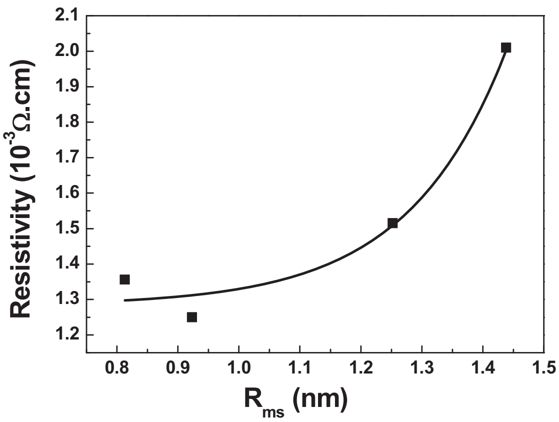


Fig.6. The relation between surface roughness and resistivity.

The optical transmission was measured as a function of wavelength from 300-800nm. Figure 7 shows the optical transmittance (T) spectra for ITO thin films versus wavelength with different Ar gas partial pressure. At the wavelength 350nm, T was quite low, with increasing wavelength to 400nm, T increased rapidly, exceeding 65%, and then became almost constant at over 92% in the 500-700nm wavelength range.

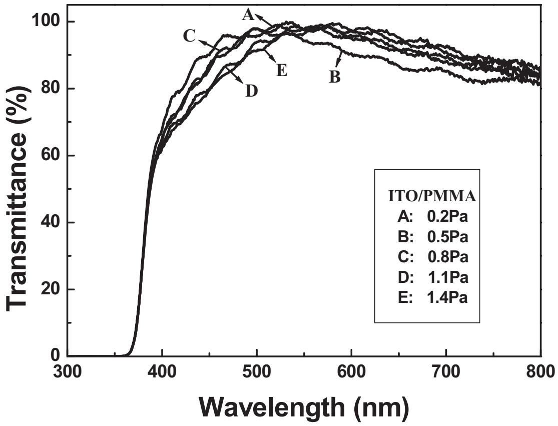


Fig.7. The optical transmittance (T) spectra for ITO thin films versus wavelength with different Ar gas pressure.

Figure 8 shows the average transmittance of ITO films prepared on PMMA substrate at different Ar gas partial pressure. The average transmittance was around 90% in the 400-760nm wavelength range. The transmission of the ITO films had no obvious changes as a function of Ar sputtering pressure. The data indicates that the average transmittance in the visible range 400-760 nm is higher for the ITO/PMMA than the ITO/PET and ITO/glass because the transmittance of the PMMA substrate is higher than that of the bare glass and PET substrate. These highly transparent ITO films make them particularly attractive application in the optical device field.

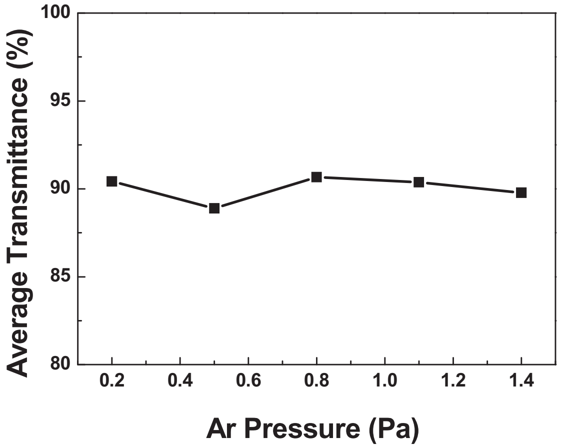


Fig.8. The average transmittance of ITO films prepared on PMMA substrate at different Ar gas partial pressure in the 400-760nm wavelength range, the average transmittance 90.42%, 88.89%, 90.67%, 90.37%, 89.78% at Ar gas partial pressure 0.2Pa, 0.5Pa, 0.8Pa, 1.1Pa and 1.4Pa, respectively.

4. Conclusions

ITO films with both high electrical conductivity and high visible transmission were obtained at Ar gas partial pressure 0.8Pa. The minimum resistivity is 1.25×10^{-3} Ω.cm and the average transmittance is around 90% in the 400-760nm wavelength range. The resistivity of ITO film increases with increasing the surface roughness, which indicates that the surface roughness plays an important role in electrical properties of the ITO films. In terms of surface roughness, resistivity and transmittance, the properties of the films prepared within Ar sputtering pressure 0.8-1.1Pa range are comparable to the best films previously reported.

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